

PATENT SPECIFICATION (11) 1 408 165

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- (21) Application No. 55218/72 (22) Filed 29 November 1972
 (31) Convention Application No. 203482 (32) Filed 30 November 1971 in
 (33) United States of America (US)
 (44) Complete Specification published 1 October 1975
 (51) INT CL² C09D 3/72 C08L 75/04
 (52) Index at acceptance
 C3R 32D11A32D12 32D1 32D6C
 32D6K 32D6L 32D9B132E3D
 32E3Y 32F2 32G2Y 32J2F
 32J2Y 32KH 32KL C11 C12
 C25 C28A C28X C9B C9N
 L2X L6G



(54) TOLUENE DIISOCYANATE URETHANE LACQUER SOLUTIONS

(71) We, WITCO CHEMICAL CORPORATION, a Corporation organised and existing under the laws of the State of Delaware, United States of America, of 277 Park Avenue, New York, State of New York, 10017, United States of America, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

The present invention relates to a novel stable, balanced, one-component chain-extended linear polyurethane lacquer solution derived specifically from toluene diisocyanate (TDI), prepared in an alcoholic solvent, which contains a C₁-C₈ monohydric alcohol at a concentration of at least 5 per cent by volume.

Polyurethane lacquer solutions generally consist of a urethane or urethane-urea type of polymer made from a difunctional polyol in compatible solvent or solvents. They are prepared by reacting a polyester or polyether with a diisocyanate to a desired free NCO value to form a prepolymer. This prepolymer is either made in a solvent or put into solution and appropriate chain-extending agents such as diamines and monoamines are added to the prepolymer solution in specific amounts to obtain a solution of desired viscosity and a desired solids content. A monohydric alcohol is sometimes added at a terminal point during the chain-extension to stop the chain growth and to achieve a desired viscosity.

U.S. 3,223,672, for instance, issued on 14 December 1965 to Schniepp, teaches the preparation of polyurethanes from substantially equimolar amounts of an aromatic diisocyanate and an aliphatic polyester in toluene at elevated temperatures, diluted to

a desired viscosity and solids content with a highly polar solvent such as methyl ethyl ketone.

U.S. 3,483,167 issued 9 December 1969 to Sommer et al, teaches viscosity control in the chain-extension of linear polyurethanes using a combination of chain-extenders consisting of certain diamines, hydrazines and hydrazides and a dihydroxy aliphatic monoamine, wherein the chain-extending reaction is stopped at the point of reaching a desired viscosity by adding a monohydric alcohol.

The present invention, provides a one component polyurethane lacquer solution which comprises a chain extended prepolymer of a linear hydroxy terminated polyether or polyester and toluene diisocyanate, the chain extension having been brought about by adding an amount of menthane diamine and/or isophorone diamine sufficient to react completely with 80 to 96 per cent of the available isocyanate groups of the prepolymer and at least where the prepolymer is derived from a polyester, an amount of one or more C₂ to C₂₂ alkyl and/or cycloalkyl monoamines sufficient to completely react with up to 10 per cent of the available isocyanate groups of the prepolymer, the resulting product being dissolved in a solvent mixture which comprises at least 5 per cent by volume of a C₁ to C₈ alkanol and such other inert organic solvents as may be required to produce a balanced solution as hereinafter defined.

By the term "balanced solution" is meant a solution in which all the components remain in solution at a desired viscosity and solids content after all the ingredients have reacted.

A commercially feasible balanced one-component lacquer solution derived from

[Price

TDI as the sole source of -NCO, wherein certain objectionable toxic and odoriferous solvents are used sparingly or not at all, was not available before this invention. These solvents can be used in higher concentrations without affecting the stability of the solution but because of their objectionable qualities, as little as possible of these solvents are employed to arrive at a stable, balanced lacquer solution.

It is surprising that the monohydric alcohol in the present lacquer solution does not inhibit the desired chain-extension of the polyurethane prepolymer with the cycloaliphatic diamine and alkyl monoamine, and that balanced solutions can be prepared over a wide range of desired viscosities and solids content. It would be expected instead, from the teachings of Sommer et al United States Specification No. 3,483,167, vide supra, that the monohydric alcohol would terminate the chain-extension reaction in the present solvent system, indeed even before it started.

The solvents used in the present invention, as mentioned above, comprises one or more C₁-C₃ monohydric alcohols which must be alkanols (i.e. methanol, ethanol, propanol, isopropanol, butanol, the isobutanols and the amyl alcohols, and mixtures thereof.) The higher alcohols are less preferred because of their slow evaporation rate. One useful combination is a mixture of methanol and isopropanol, a 5 to 15 per cent by volume solution of methanol in isopropanol being particularly useful as a solvent for polyether derived polyurethane lacquer solutions. Methanol is not preferred, however, in the solvent system wherein the polyurethane prepolymer is derived from a polyester, because of the problem of short term stability of the resultant lacquer solution at room temperature.

In addition to the alcohol, the solvent system usually includes conventional inert solvents such as toluene, benzene, xylene, methylethyl ketone, methyl butyl ketone, ethyl acetate, methyl isobutyl acetate, cyclohexanone, and mixtures, and in general any inert lacquer solvent well known in the art may be used. It is an advantage of the present invention that certain odoriferous, toxic and expensive solvents such as dimethylformamide, dimethylsulfoxide, and tetrahydrofuran, for instance, are used sparingly i.e. between 0 to 20 per cent by volume, as needed to produce a balanced solution. Solvents such as toluene and methylethyl ketone are readily available, are of low cost, and are preferred in the present alcoholic solvent system, but a minor amount of other solvent such as dimethylformamide may be required to arrive at a fully balanced system particularly when using polyester derived polyurethane prepolymer. The solution gen-

erally remains stable during ordinary shelf life and, upon evaporation of the solvent, leaves an unblemished film having good stress-strain properties.

In the preparation of linear polyurethane lacquer solutions from hydroxy-terminated polyethers by the present invention, substantially all of the solvent system can be a C₁-C₃ monohydric alcohol or mixtures of alcohols. With lacquer solutions of polymers prepared from hydroxy-terminated polyesters, another lacquer solvent is usually required and the C₁-C₃ alcohol may be present in amounts of as little as 5 to 10 per cent and up to 80 per cent by volume. As stated above, the conventional inert lacquer solvents known in the art are employed to dilute the monohydric alcohol. Concentrations of monohydric alcohol of less than 5 per cent by volume are not preferred because of the danger of loss of solution balance as the volume of alcohol approaches 5 per cent and less. The use of objectionable solvents with the monohydric alcohol, as mentioned above, is kept to a minimum, i.e. sufficient to arrive at a solvent system wherein the growing polymer remains soluble and stable. In some cases, little, if any, objectionable solvent need be added at all, as for instance in systems having concentrations of aliphatic monoamine in the upper range, i.e. 8 to 10 per cent, based on available -NCO, and in systems where a polyether prepolymer is employed to render a balanced lacquer solution.

The prepolymer in the present lacquer solutions is prepared as known in the art by reacting an excess of TDI, which can be the 2,4- or 2,6-isomer or mixtures thereof, with a linear hydroxy-terminated polyether or polyester. The -NCO/OH ratio of the reactants producing the present prepolymer may vary from 1.2/1 to 2.0/1 to give good results. It is surprising that films produced, particularly when using prepolymers from TDI and a polyester, exhibit good resistance to discolouring and to loss in tensile strength in ultraviolet light. Appreciably branched polyethers and polyesters are not included in the scope of the present invention because they tend to form gelled products.

Only prepolymers derived substantially from TDI are employed in the present invention. Although other aromatic diisocyanates may be compatible in the present solvent system, they do not render films which resist breakdown by ultraviolet light, even with added antioxidants and U.V. absorbers. It is surprising that only the present chain-extended linear polyurethane lacquer solutions derived from TDI render films having these beneficial properties. Distinctly minor quantities, for instance up to 3 per cent by weight, of other aromatic diisocyanates, although not preferred, may be included in

the present system.

It is preferred to use a 80-20 isomeric mixture of 2,4- and 2,6-isomers of TDI, respectively, because of the commercially availability of this isomeric mixture and its low cost. However, the 2,4-isomer presents fewer problems and is also preferred, and the commercial available 65-35 isomeric mixture can also be used effectively in the present system.

Representative examples of hydroxy-terminated polyesters which are reacted with TDI to prepare the prepolymers of the present lacquer solutions are the condensation products of an aliphatic dicarboxylic acid, or mixture of aliphatic and aromatic dicarboxylic acids, with a glycol. Suitable glycols include ethylene glycol, the propylene glycols, the butylene glycols, 1, 6 hexane diol, and decamethylene glycol.

Representative examples of aliphatic dicarboxylic acids are succinic acid, glutaric acid, adipic acid, azelaic acid, sebacic acid. Adipic acid, being readily available and of low cost, is preferred. The aliphatic acid may be mixed with a quantity of aromatic dicarboxylic acid such as phthalic acid to produce lacquers which render films of greater hardness.

Representative examples of hydroxy-terminated polyethers useful in preparing the prepolymers with TDI of the present lacquer solutions are the reaction product of an epoxide and a diol. Generally epichlorohydrin or any 1,2-alkylene oxide having from 2 to 10 carbon atoms, and preferably from 2 to 4 carbon atoms may be used, to prepare polyethers such as poly(oxyethylene)glycol, poly(1,2-oxypropylene)glycol, poly(oxytetramethylene)glycol, and other like polyethers as known in the art.

The hydroxy-terminated polyether or polyester used in the present process must be substantially anhydrous. This result may be achieved by any means well known in the art such as heating the diol under vacuum or heating the solution of diol in a common aromatic hydrocarbon solvent such as benzene, to form an azeotrope which distills therefrom. After drying, the residual aromatic solvent can remain as part of the solvent system of the final lacquer solution.

The amine chain extenders, according to the present invention, are cycloaliphatic diamines which may be either menthane diamine or isophorone diamine and mixtures thereof, and an alkyl monoamine having alkyl chains of from 2 to 22 carbon atoms, which can be a primary or a secondary alkyl or cycloalkyl amine such as ethylamine, diethylamine, propylamine, dipropylamine, butylamine, isobutylamine, cyclohexylamine, dodecylamine, didodecylamine, hexadecylamine, or octadecylamine, and mixtures thereof, the cycloaliphatic diamine

being present in concentrations of 80 to 96 per cent based on the available -NCO, and the monoamine up to 10 per cent. Higher or lower viscosity lacquer solutions are prepared depending on the quantity of cycloaliphatic diamine in the polymer, as known in the art. With polyurethane lacquer solutions derived from polyether polyurethane prepolymers, as described above, little or no alkyl monoamine is required to produce a balanced stable solution. However, when polyester polyurethane prepolymers are employed, a quantity of alkyl monoamine is required, as stated above, dependent upon the desired viscosity and solids content of the final lacquer solution and the desired characteristics of the film to be made thereby. Quantities of somewhat less than 80 per cent or somewhat more than 96 per cent of cycloaliphatic diamine, and somewhat more than 10 per cent monoamine may be employed in the present system, but are not preferred.

It is surprising that various other diamines both aliphatic and aromatic are not operative in the present lacquer solutions and that various aromatic monoamines which were tested are also inoperative. Only the specific cycloaliphatic diamines as described above, and only alkyl monoamines, have been found to be operable in the present polyurethane lacquer solution system, and in the proportions set forth elsewhere herein, to render stable balanced solutions which yield films and coatings having good tensile strength.

A storage-stable one-component balanced linear polyurethane lacquer solution generally having a total solids content of up to 50 per cent, by weight (e.g. 15 to 50 per cent) and a viscosity which may be as high as 90,000 cps or more (e.g. up to 100,000 cps) at 25°C., is made possible by the present invention, from which unblemished films and coatings having good tensile strength are made by evaporation of the solvent therefrom. The stress-strain properties of films prepared from the present lacquer solutions are equal to or better than films produced by the prior art methods and have the advantage of being more economical and made with readily available ingredients, as described hereinbefore.

Films and coatings prepared from a polyester based prepolymer have the added advantage of having good resistance to ultraviolet light. However, those produced from polyether based prepolymers tend to break down under U.V. light and the lacquer solutions from which these films are made are treated with small amounts of antioxidants and U.V. absorbers (about 1 per cent by weight) if the film or coating is to be protected from attack by ultraviolet light. Antioxidants such as the dibutylated phenols (Ionol, Shell Chemical Company) and U.V.

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absorbers such as the benzotriazoles (TINUVIN — 328, Geigy Ciba Corporation) are well known in the art and require no further discussion herein. (IONOL and TINUVIN are registered Trade Marks.) Of course, the non-staining type of antioxidants such as 2,6-ditertiarybutyl-4-methyl phenol are preferred.

There is no need for laborious heating, refluxing or autoclaving in the preparation of the present lacquer solutions; the simple rapid admixture, usually admixed in less than five minutes, with agitation, yields a product on standing, which is the final lacquer solution to be used for its ultimate purpose. It is an advantage in preparing the present lacquer solutions that there is no visual endpoint to be observed. The endpoint is pre-set by calculation of the solids content and viscosity based on the quantity of cycloaliphatic diamine, alkyl monoamine and prepolymer in the alcoholic solvent system from which an unblemished film is prepared having good tensile strength.

Polyurethane films prepared from the polyurethane lacquer solutions of the present invention have the added benefit of improved hand and better nonblocking properties, when the prepolymer chain-extension is accomplished by using a fatty monoamine such as dodecylamine, hexadecylamine, and octadecylamine, or branched chain fatty amine. These beneficial results are also obtained with mixtures of lower alkyl monoamines and fatty monoamines and these mixtures are included in the scope of the present invention.

In summary, the present invention makes available an inexpensive, commercially feasible, stable, balanced one-component, linear polyurethane lacquer solution of controlled viscosity derived from a toluene diisocyanate. All the ingredients of the lacquer solution, the prepolymer, the cycloaliphatic diamine and the alkyl monoamine, in the proportions stated, react at just the correct rate in the particular alcohol solvent system so that the growing polymer remains soluble and stable, and ready for use when needed to make films, layers and coatings, by pouring out the lacquer solution and evaporating the solvent.

The following examples are intended to illustrate the present invention in specific aspects and are not intended to be limiting of the scope thereof in any way.

In the examples the term "parts" means parts by weight except where otherwise stated.

Example 1

One thousand parts of a polyester of adipic acid and ethylene glycol (hydroxyl number 112) are dehydrated at about 120°C. for about 30 minutes at 10 mm Hg, and then

allowed to react for about 1½ hours at 105°C. with 300 parts of toluene diisocyanate (80 per cent 2,4- and 20 per cent 2,6-isomer by weight ratio). The prepolymer has a free -NCO value of 4.6. Enough methylethyl ketone is added to make a 75 per cent by weight solution. Additional methylethyl ketone may be added at this time or any later time for purposes of arriving at a desired viscosity. With mixing, the following solution is added over a period of about five minutes: isophorone diamine, monobutylamine in ethanol and dimethylformamide. The isophorone diamine quantity is sufficient to react with about 92 per cent of the free available -NCO value and the monobutylamine with about 5 per cent of the free available -NCO value. Solvents are in a ratio of 80 per cent methylethyl ketone, 15 per cent ethanol and 5 per cent dimethylformamide by volume. The final solution has a total solids content of 33 per cent by weight and viscosity of 27,000 cps at 25°C.

Example 2

One thousand parts of poly(tetramethylene adipate) having a hydroxyl number 135, acid number of 0.3, are dehydrated at about 110°C. for 30 minutes at 2 mm Hg and then allowed to react for about 1½ hours at 105°C. with 400 parts of 2,4-toluene diisocyanate. The prepolymer has a free -NCO value of 6.45. Enough toluene is added to make a 75 per cent by weight solution. Enough additional toluene is added to the prepolymer to bring the weight to approximately 50 per cent of the final weight of the lacquer solution. To another vessel is added a solution of menthane diamine and diethylamine in the remainder of the toluene, isopropanol and dimethylformamide. The contents of each container is fed through proportionating pumps to a mixing head and fed to a receiving container. The menthane diamine is sufficient to react with about 94 per cent of the available -NCO value and the diethylamine with 2 per cent of the free available -NCO value. The solvents are in the ratio of 65 per cent toluene, 20 per cent isopropanol and 15 per cent dimethylformamide by volume. The final solution has a total solids content of 25 per cent by weight and viscosity of 15,000 cps at 25°C.

Example 3

A three mil film prepared by casting the product of Example 2 on release paper, evaporating the solvent and drying the film has a tensile strength of over 7,000 psi, elongation of about 480 per cent, Die C tear strength of about 475 pli and split (trouser) tear of about 350 pli. The clear film shows only slight discoloration under ultraviolet light for up to about 100 hours, and the tensile strength shows only a slight drop of from

6,000 to 4,000 psi after exposure in an Atlas Weatherometer at 145°F. and 65 per cent relative humidity for about 100 hours. (Atlas is a registered Trade Mark.)

Example 4

One thousand parts of poly(1,2-oxypropylene)glycol having a hydroxyl number of 160 are dehydrated at about 120°C. for 30 minutes at 5 mm Hg and then allowed to react for about two hours at 110°C. with 385 parts of toluene diisocyanate, 65 per cent 2,4- 35 per cent 2,6-isomer. The prepolymer has a free -NCO value 4.8. Sufficient toluene is added to make a 75 per cent by weight solution. To the above prepolymer in toluene is added the remainder of the toluene to be used. With good mixing the following solution is added over a period of 2 minutes: isophorone diamine and dibutylamine in a solvent system of isopropanol-dimethyl formamide. The diamine quantity is sufficient to react with 90 per cent of the free available -NCO value and the monoamine with about 7 per cent of the free available -NCO value. The final solution has a total solids content of 35 per cent by weight and a viscosity of 17,000 cps at 25°C.

Example 5

One thousand parts of poly(oxytetramethylene)glycol are dehydrated at about 100°C. for about one hour at about 1 mm Hg, and then allowed to react for about one hour at 100°C. with 300 parts of 2,4-toluene diisocyanate. The prepolymer has a free NCO value of 4.6. Enough methylethyl ketone (MEK) is added to make a 50 per cent by weight solution. To the above prepolymer solution is added the remainder of the MEK as needed. With mixing a solution of isophorone diamine and n-octadecylamine in ethanol is added over a period of 1 minute. The diamine is sufficient to react with about 91 per cent of the free available -NCO value and the monoamine with 6 per cent of the -NCO value. The solvents are in the ratio of 80 per cent MEK and 20 per cent ethanol by volume. The final solution has a total solids content of 35 per cent by weight and viscosity of 9,000 cps at 25°C.

Example 6

One thousand parts of poly(1,6-hexane adipate) having a hydroxyl number of 230 are dehydrated at 100°C. for one hour, at 1 mm Hg. To this is added 606 parts of toluene diisocyanate, 80 per cent 2,4- and 20 per cent 2,6-isomer, and allowed to react for two hours at 100°C. This prepolymer has a free NCO value of 7.5. Toluene is added to make a 75 per cent by weight solution. To this prepolymer solution additional toluene and dimethylformamide are added. With good

mixing is added a solution of isophorone diamine and n-propylamine in n-propanol, in one minute. The isophorone diamine will react with the 94 per cent of the free -NCO and the monoamine with 5 per cent. The solvents are 70 per cent toluene, 20 per cent n-propanol and 10 per cent dimethylformamide by volume. The final solution has a total solids content of 25 per cent by weight and a viscosity of 11,000 cps at 25°C.

Example 7

One thousand parts of poly(ethylene adipate) having a hydroxyl number of 112 and dehydrated at 100°C. for ½ hour at 1 mm Hg and then added to 312 parts of 2,4 toluene diisocyanate and allowed to react for 1½ hours at 100°C. This prepolymer has NCO value of 5.10. Methylethyl ketone is added to make 75 per cent by weight solution. To this solution of prepolymer are added the remaining methylethyl ketone and the required acetone and dimethylformamide. A solution of isophorone diamine, and octadecylamine in isopropanol is added with good mixing in 1½ minutes. About 7 per cent of the free -NCO reacts with the octadecylamine and 92 per cent with the isophorone diamine. The solvents are 40 per cent methylethyl ketone, 40 per cent acetone, 10 per cent isopropanol and 10 per cent dimethylformamide by volume. The total solids content of the final solution are 25 per cent by weight and the viscosity is 10,000 cps at 25°C.

A film prepared using this lacquer solution has better feel and non-blocking properties than one prepared from a similar lacquer solution wherein the octadecylamine is replaced with butylamine.

Example 8

One thousand parts of poly(ethylene/propylene adipate) having a hydroxyl number of 60 are dehydrated at 100°C. for ½ hour at 1 mm Hg. 165 parts of toluene diisocyanate, 80 per cent 2,4- 20 per cent 2,6-isomer, are added and allowed to react at 90°C. for 3 hours. This prepolymer has a free -NCO value of 3.0. The required amount of toluene is added to the prepolymer. A solution of isophorone diamine and diethylamine in isopropanol and dimethylformamide is added with good mixing to the prepolymer toluene solution in 1 minute. The isophorone diamine reacts with 95 per cent of the free -NCO and the monoamine with 3 per cent of the free NCO. The solvent comprises 70 per cent toluene, 25 per cent isopropanol, and 5 per cent dimethylformamide by volume. The final solution has a total solids content of 35 per cent by weight and a viscosity of 50,000 cps at 25°C.

Example 9

One thousand parts of poly(1,2-oxypropylene)glycol having a hydroxyl number of 142.0 are dehydrated at 1 mm Hg and then allowed to react with 439 parts 5 of toluene diisocyanate (80 per cent 2,4- and 20 per cent 2,6-) for two hours at 100°C. The prepolymer has a free -NCO value of 5.75. The prepolymer is added with good mixing to 10 a solution of isophorone diamine and diethylamine in an alcoholic solution comprising 5 per cent by volume methanol in isopropanol in quantities sufficient to react 96 per cent of the free -NCO with the diamine and 3 per 15 cent with the monoamine. The final lacquer solution contains 35 per cent by weight total solids, and has a viscosity of about 15,000 cps at 25°C.

WHAT WE CLAIM IS:—

1. A one component polyurethane lacquer solution which comprises a chain extended prepolymer of a linear hydroxy terminated polyether or polyester and 25 toluene diisocyanate, the chain extension having been brought about by adding an amount of menthane diamine and/or isophorone diamine sufficient to react completely with 80 to 96 per cent of the available isocyanate groups of the prepolymer and at 30 least, where the prepolymer is derived from a polyester, an amount of one or more C₂ to C₂₂ alkyl and/or cycloalkyl monoamines sufficient to completely react with up to 10 per cent of the available isocyanate groups of the prepolymer, the resulting product being dissolved in a solvent mixture which comprises at least 5 per cent by volume of C₁ to C₃ alkanol and such other inert organic solvents 40 as may be required to produce a balanced solution as hereinbefore defined.

2. A composition as claimed in claim 1 wherein the prepolymer is one in which the ratio of isocyanate groups to hydroxy groups in the reactants from which the prepolymer is 45 formed is in the range from 1.2:1 to 2:1.

3. A composition as claimed in claim 1 or claim 2 wherein the prepolymer is derived from 2,4-toluenediisocyanate, a mixture of 50 80 per cent by weight of 2,4-toluenediisocyanate and 20 per cent by weight of 2,6-toluenediisocyanate or a mixture of 65 per cent by weight of 2,4-toluenediisocyanate and 35 per cent by 55 weight of 2,6-toluenediisocyanate.

4. A composition as claimed in any one of claims 1 to 3 wherein the prepolymer is derived from a toluene diisocyanate and a polyester which is the condensation product 60 of a polyol with an aliphatic dicarboxylic acid or a mixture thereof with an aromatic dicarboxylic acid.

5. A composition as claimed in claim 4 wherein the polyol is ethylene glycol, a propylene glycol, a butylene glycol, 1,6-hexane 65

diol or decamethylene glycol.

6. A composition as claimed in claim 4 or claim 5 wherein the aliphatic dicarboxylic acid is succinic acid, glutaric acid, adipic acid, azelaic acid or sebacic acid. 70

7. A composition as claimed in any one of claims 4 to 6 wherein the aromatic dicarboxylic acid is phthalic acid.

8. A composition as claimed in any one of claims 1 to 3 wherein the prepolymer is derived from a toluene diisocyanate and a polyether which is the reaction product of an epoxide and a diol. 75

9. A composition as claimed in claim 8 wherein the epoxide is a 1,2-alkylene oxide having from 2 to 10 carbon atoms. 80

10. A composition as claimed in claim 9 wherein the 1,2-alkylene oxide contains from 2 to 4 carbon atoms.

11. A composition as claimed in any one of claims 8 to 10 wherein the polyether is poly(oxyethylene)glycol, poly(1,2-oxypropylene)glycol or poly(oxytetramethylene)glycol. 85

12. A composition as claimed in any preceding claim wherein the alkyl or cycloalkyl monoamine is a primary or secondary amine. 90

13. A composition as claimed in any preceding claim wherein the alkyl monoamine is ethylamine, diethylamine, propylamine, dipropylamine, butylamine, isobutylamine, dodecylamine, didodecylamine, hexadecylamine or octadecylamine. 95

14. A composition as claimed in claim 12 wherein the cycloalkyl monoamine is cyclohexylamine. 100

15. A composition as claimed in any preceding claim wherein the solvent is a mixture in which the alkanol is present in an amount of from 5 to 10 per cent by volume. 105

16. A composition as claimed in any preceding claim wherein the solvent is a mixture which comprises methanol and isopropanol.

17. A composition as claimed in claim 16 wherein the mixture contains from 5 to 15 per cent by volume methanol dissolved in isopropanol. 110

18. A composition as claimed in any preceding claim wherein the solvent includes one or more of toluene, benzene, xylene, methylethyl ketone, methyl butyl ketone, ethyl acetate, methyl isobutyl acetate and cyclohexanone. 115

19. A composition as claimed in any preceding claim wherein the solvent is a mixture which comprises ethanol and methylethyl ketone. 120

20. A composition as claimed in any preceding claim wherein the solvent is a mixture which comprises isopropanol and toluene. 125

21. A composition as claimed in claim 1 and substantially as hereinbefore described in any one of the foregoing specific Examples.

22. A process for the production of a one 130

- component polyurethane lacquer solution which process comprises reacting a toluene diisocyanate with a linear hydroxy terminated polyether or polyester to yield a prepolymer, chain extending the said prepolymer in a solvent mixture comprising at least 5 per cent by volume of a C₁ to C₅ alkanol by adding an amount of menthane diamine and/or isophorone diamine sufficient to react completely with 80 to 96 per cent of the available isocyanate groups of the prepolymer and, at least where the prepolymer is derived from a polyester, an amount of one or more C₂ to C₂₂ alkyl and/or cycloalkyl monoamines sufficient to react completely with up to 10 per cent of the available isocyanate groups of the prepolymer, and during or after the chain extension reaction adding further inert organic solvent(s) or C₁ to C₅ alkanol(s) as required to produce a balanced solution as hereinbefore defined in which one or more C₁ to C₅ alkanols constitute at least 5 per cent of the solvent mixture.
23. A process as claimed in claim 22 wherein the solvent comprises a mixture of C₁ to C₅ alkanol and methylethyl ketone.
24. A process as claimed in claim 22 wherein the solvent comprises a mixture of C₁ to C₅ alkanol and toluene.
25. A process as claimed in claim 22 wherein the solvent comprises ethanol and methylethyl ketone.
26. A process as claimed in claim 22 wherein the solvent comprises isopropanol and toluene.
27. A process as claimed in claim 22 where the prepolymer is derived from a polyether and wherein the solvent consists of a mixture of from 5 to 15 per cent by volume methanol in isopropanol.
28. A process as claimed in claim 22 and substantially as hereinbefore described in any one of the foregoing specific Examples.
29. A polyurethane lacquer whenever obtained by a process as claimed in any one of claims 22 to 28.

BOULT, WADE & TENNANT
Chartered Patent Agents,
34 Cursitor Street,
London EC4A 1PQ